

# Science of Geological Carbon Sequestration

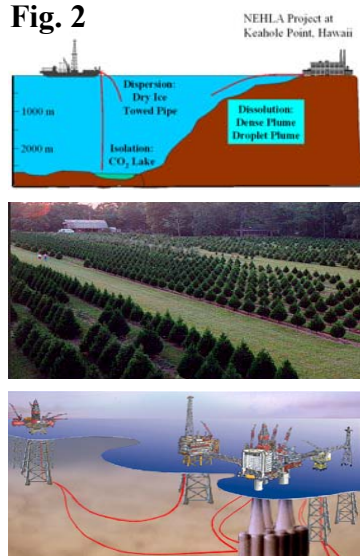
## Integration of Experimentation and Simulation

### Introduction

The Intergovernmental Panel on Climate Change (IPCC) predicted in its 1995 “business as usual” energy scenario that the global emissions of CO<sub>2</sub> to the atmosphere would increase from 7.4GtC (Giga tons, or billion tons, of carbon) per year in 1997 to 26GtC/yr by 2100, resulting in a doubling of atmospheric CO<sub>2</sub> concentration by the middle of this century. There is consensus in the scientific community (e.g., *Science*<sup>[1]</sup>) that increased levels of greenhouse gases such as CO<sub>2</sub> are adversely affecting the global environment as evidenced by recent trends in global warming and dramatic changes in weather patterns.



**Fig. 2**



In February 2002, President Bush announced the Global Climate Change Initiative, committing the nation to cut greenhouse gas intensity (the ratio of greenhouse gas emissions to economic output) by 18% over the next 10 yrs. To achieve this ambitious goal, the President ordered the development of a National Climate Change Technology Initiative (NCCTI) to pursue advanced, cost-effective technologies.

There are three major carbon sequestration options currently under DOE evaluation (Fig. 2): Sequestration in the ocean, in terrestrial ecosystems, and in geologic formations. Although the ocean represents the largest potential sink for CO<sub>2</sub>, this option is the least understood in terms of its sequestration mechanisms and environmental impacts. Terrestrial sequestration is achieved through enhanced natural processes and is hence desirable. However, the total capacity of this option is estimated to be small (1.5-2.0GtC).

Disposal of CO<sub>2</sub> in geologic formations represents the most promising near-term solution to the problem of long-term carbon sequestration<sup>[2]</sup>. The reasons are as follows: 1) The global capacity of geologic formations is large enough to store many decades or centuries worth of emissions. Table 1 shows some conservative estimates of CO<sub>2</sub> storage of domestic geologic formations. For example, the storage capacity in saline formations is estimated to be 5-500Gt in the U.S. and 320-10,000Gt of CO<sub>2</sub> globally. 2) Geologic formations are widely available and in close proximity to power generation plants, from which one third of U.S. CO<sub>2</sub> emissions come. There are about 600 power plants sitting above deep saline reservoirs and 32 plants within 50 miles of existing or planned enhanced oil recovery (EOR) fields. 3) The geologic formations have a proven record for storing oil, gas, coal bed methane, or natural CO<sub>2</sub> over geologic time periods. 4) The petroleum industry has already developed the necessary technology for injecting CO<sub>2</sub> in these reservoirs through its

**Table 1. CO<sub>2</sub> Storage Capacities of Domestic Geologic Formations**

	Estimated CO <sub>2</sub> Storage Capacity (Gt of CO <sub>2</sub> )
Unmineable Coal Beds	15-20
Depleting Oil Reservoirs	40-50
Depleting Gas Reservoirs	80-100
Saline Formations	5-500
High Organic Shales	TBD
<b>TOTAL</b>	<b>140-670</b>

EOR program. 5) Enhanced oil, gas, or methane production from the respective oil, gas reservoirs, or coal beds due to injected CO<sub>2</sub> provides an economical incentive to partially offset the sequestration cost.

This proposed LDRD/DR research focuses on fundamental issues associated with sequestering CO<sub>2</sub> in depleted oil reservoirs and saline (brine) formations. Oil reservoirs are estimated to have capacity to store several years worth of CO<sub>2</sub> emissions. The necessary surface and downhole infrastructure is already in place for injecting CO<sub>2</sub> into depleted oil reservoirs. If not damaged during production, the structures of oil reservoirs that trapped oil for a geologic timeframe may also be capable of trapping CO<sub>2</sub>. Therefore, sequestration in depleted oil reservoirs may be the nearest-term, cost effective option to be deployed. In spite of limited knowledge, injection of CO<sub>2</sub> into saline formations represents a sequestration option of large potential capacity (Table 1, 500 GtC). Sequestration in oil reservoirs and saline formations is schematically illustrated in Fig.

3. In these formations, the main CO<sub>2</sub> sequestration mechanisms are structural trapping (due to low-permeability traps), solubility trapping (scCO<sub>2</sub> dissolution into oil and water), and mineral trapping (reactions with minerals to form new, permanent mineral products). Viscous fingering, gravity segregation, miscibility, reaction kinetics, and possible leakage through natural/artificial pathways are some of the factors that may significantly affect the sequestration capacity as well as the fate and redistribution of injected CO<sub>2</sub>.

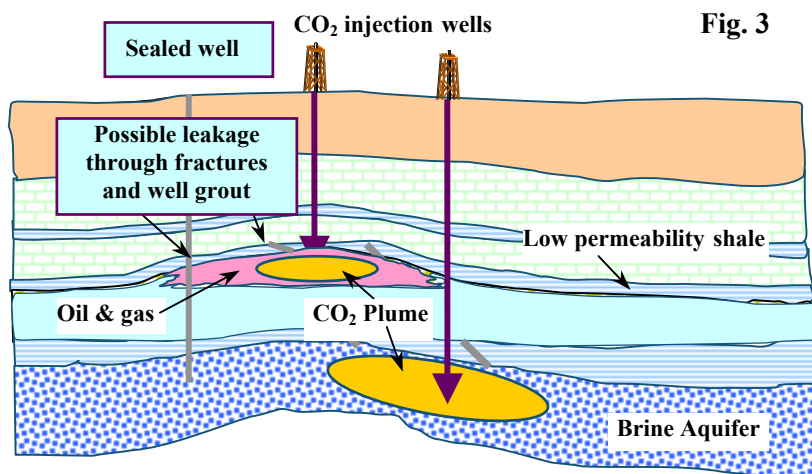


Fig. 3

### Outstanding Scientific Issues

The primary scientific issues for CO<sub>2</sub> sequestration are significantly different than for CO<sub>2</sub> flooding in EOR operations because of the large contrast in timeframes: several yrs (for flooding) vs. 100-1000s of yrs (for sequestration). For instance, a leakage of 1% per year may not be a concern for CO<sub>2</sub> flooding, but it is unacceptable over 100s of yrs for CO<sub>2</sub> sequestration. The key research issues related to the viability of geologic sequestration revolve around the ultimate fate of the injected CO<sub>2</sub> and include the following:

1. Can the leakage rates from a CO<sub>2</sub> storage reservoir and their potential impact be estimated? Slow leakage of CO<sub>2</sub> to overlying subsurface formations may be inconsequential or even beneficial (e.g., if to other deep saline aquifers). On the other hand, slow leakage to the atmosphere would be unacceptable. A highly accurate reservoir-scale numerical model that incorporates all of the primary flow, transport, thermodynamics and chemical reactions is required to evaluate such leakage scenarios.
2. What are the relative permeability relationships among scCO<sub>2</sub>, brine, and oil? At reservoir pressure and temperature, CO<sub>2</sub> will be injected as a supercritical (scCO<sub>2</sub>) fluid (critical point = 31°C and 74 bars) that is immiscible with both brine and oil, leading to complex two- and

three-phase permeability relations that are essential to understand and predict multi-phase flow but are not well reported in literature.

3. How is the host environment (sandstone or limestone reservoirs) affected by injection of scCO<sub>2</sub> (changes in porosity, permeability, and mineralogy)? Dissolution of scCO<sub>2</sub> into brine creates an acidic solution that will be out of equilibrium with the host rock. For example, Fig. 4 shows the formation of wormholes in limestone cores resulting from brine-CO<sub>2</sub> injection in a flow-through experiment conducted by our external collaborators at NM Tech. Innovative experimental and simulation approaches are needed to understand and predict the formation of wormholes. More generally, under what conditions do the wormholes occur and how do they develop? How does dissolution-precipitation affect reservoir permeability and porosity and hence the injectivity and the sequestration capacity of the reservoir? What numerical methods are required to simulate phenomena such as wormhole formation in two- or three-phase fluid environments?
4. What are the predominant reactions and reaction rates at the reservoir conditions? Does CO<sub>2</sub> react with the rock matrix at a reasonable rate to permanently trap the CO<sub>2</sub>? Do the reactions plug the preferred flow paths, especially fractures? Reactions among multi-phase fluids, carbonate and silicates are



Fig. 4 Limestone after extensive CO<sub>2</sub> and brine injection <sup>[3]</sup>. Note the existence of wormholes.

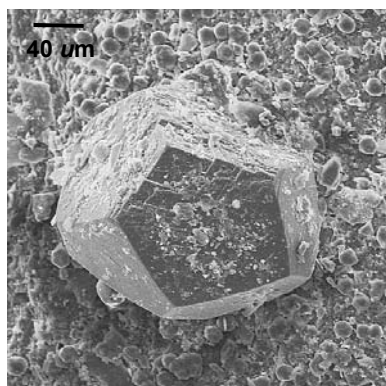


Fig. 5 Precipitation of siderite and analcime (small, rounded grains) on shale in supercritical CO<sub>2</sub>-brine-rock experiment. Siderite and analcime did not grow on aquifer materials.

intimately coupled <sup>[4]</sup> and not as simple as has been depicted in the literature. A combination of flow-through and batch experiments will be required to determine *effective* reaction rates required for numerical models for geochemical reactions in CO<sub>2</sub> reservoirs.

5. How permeable are the materials (shale and cement grout) that trap the CO<sub>2</sub> in the reservoir? Can scCO<sub>2</sub> negatively modify these sealing materials? Our experimental work shows that the mutual solubility of scCO<sub>2</sub> and H<sub>2</sub>O enhances chemical reaction with caprock materials (Fig. 5). Such chemical reactions, depending on volume changes and material transport, may either introduce higher permeability channels or effectively seal existing pathways for CO<sub>2</sub> migration.
6. How certain are the answers to these questions? Confidence

will be built by application of numerical models for reservoir performance and formal uncertainty quantification approaches to simulation of CO<sub>2</sub> injection sites (short-term behavior, e.g., Hobbs, NM) and natural analog sites (long-term behavior, e.g., Bravo Dome, NM).

## Science and Technology Objectives

Effective sequestration of CO<sub>2</sub> and assurance of public safety from escaping CO<sub>2</sub> require accurate prediction of long-term behavior of CO<sub>2</sub> in the subsurface. Our proposal will provide

the experimental data and numerical codes necessary to calculate the long-term fate of CO<sub>2</sub> after injection into depleted oil or saline/brine reservoirs. To arrive at this objective, our proposal will focus on the key technical and knowledge barriers to understanding the migration and storage of CO<sub>2</sub> in typical reservoir environments.

The injection of scCO<sub>2</sub> starts in a reservoir with readily characterized permeability, porosity, fluid-phase distribution (brine or brine + oil), and mineralogy. The unknown is how the reservoir responds to CO<sub>2</sub> in terms of changes in these basic properties. Our experimental approach will use dynamic flow-through and static batch methods at the core-scale to derive relative permeability relations for CO<sub>2</sub>, oil, and brine with an emphasis on identifying heterogeneous behavior such as viscous fingering, wormhole formation, or flow-path partitioning of fluids. The experiments will yield fundamental data on mixed fluid-rock reactions and on the role of mineral precipitation and dissolution in modifications to permeability and porosity as well as in the permanent sequestration of CO<sub>2</sub> as precipitated carbonates. We will also determine the permeability and reactivity of scCO<sub>2</sub> to the caprock (shale) that provides a barrier to CO<sub>2</sub> migration as well as to the cement grout that seals the wells penetrating this caprock. These measurements will provide the basis for calculations of the long-term integrity of the reservoirs.

The experimental data will be integrated into a two-level simulation approach that will be used to study the interaction of scCO<sub>2</sub> with geologic media, the long-term fate, and the potential impact of failures. Experimental analyses of these long-term behaviors are not possible with relatively short-term laboratory experiments. They can however be studied effectively with comprehensive numerical models that incorporate the underlying physics that is identified through detailed experiments. Pore network and lattice Boltzmann (LB) models will incorporate the microscopic experimental results to simulate interactions at the pore scale and to develop constitutive relationships to use for modeling at the macroscopic scale. At macroscopic scales, coupled hydrodynamic, thermodynamic, and geochemical simulations will be used to study migration of CO<sub>2</sub> and its interaction with the host media when large amounts of CO<sub>2</sub> are injected. Numerical models will be built on existing LANL codes that represent the state of the art and are extensively used in the Yucca Mountain and Hanford Site Projects to study underground nuclear waste storage. The models will be refined with the data from macroscopic experiments and will incorporate the effect of parameter uncertainties (Zhang <sup>[5]</sup>) on the long-term environmental impact of disposed CO<sub>2</sub>. Integrated experiments and simulation will cover the behavior of scCO<sub>2</sub> in the most common rocks and fluid systems present in depleted oil reservoirs and saline aquifers (sandstones, carbonates, and shales with brine or brine plus oil, Table 2).

Table 2. Experiment and Simulation Matrix.

<i>Focus</i>	<b>Experiments</b>		<b>Simulation</b>
	Rocks/Materials	Fluids	
<b>Reservoir</b>	Sandstone - quartz-rich or feldspar-rich - calcite or silica cement	2-Phase: scCO <sub>2</sub> -brine 3-Phase: scCO <sub>2</sub> -brine-oil	Pore-scale - pore network - Lattice Boltzmann - Mineral reactions
	Carbonate - limestone - dolomite		Core-scale - Hybrid model
<b>CO<sub>2</sub> Traps</b>	Shale (caprock)	2-Phase: scCO <sub>2</sub> -brine	Reservoir-scale
	Cement grout		- Continuum

The experimental and modeling results will be validated by applying the two-level simulation approach to field sites: 1) The Hobbs, NM CO<sub>2</sub> injection experiment; 2) large-scale

CO<sub>2</sub> EOR field sites where data are available; and 3) Bravo Dome, NM, a naturally-occurring CO<sub>2</sub> reservoir. The CO<sub>2</sub> injection sites will test the model's short-term predictive capability (e.g., time-dependence of injection pressure, fluid migration, etc.). The Bravo Dome site will test the model's long-term predictive capability (e.g., reservoir mineralogical changes, leak rates, etc.). The Hobbs injection experiment is the first U.S. field CO<sub>2</sub> sequestration demonstration project funded during FY00-03 through DOE; LANL is one of the lead organizations<sup>[6]</sup>. While this project focuses on field injection and monitoring the migration of injected CO<sub>2</sub>, it provides a valuable data set for calibrating and validating the models to be developed in this proposed DR. Bravo Dome, NM is the world's largest and purest known natural CO<sub>2</sub> reservoir. The reservoir is mainly in sandstone, one of the primary formations considered for geologic sequestration of CO<sub>2</sub>. The field has been in operation extracting CO<sub>2</sub> for commercial uses since 1931 and has more than 300 exploration/production wells<sup>[7]</sup>. A large amount of data, such as formation properties and water chemistry, have been filed with NM Environmental Department and are thus available to us. We will also have access to some actual cores and fluid samples, on the basis of which we will perform detailed measurements, analyses, and simulations. This natural reservoir provides a unique window for understanding CO<sub>2</sub> behavior for a geologic timeframe and its long-term impacts to the host environment.

We anticipate that our innovative, integrated experimental and modeling approach will yield the most accurate and robust numerical analysis of CO<sub>2</sub> repository performance. Our experimental focus on actual measurement of CO<sub>2</sub> injected into brine- or oil + brine-saturated cores will provide a unique set of observational benchmarks to guide model development. Our two-level simulation approach will give us the required ability to incorporate the effect of microscopic processes into a reservoir-scale flow and reactive transport model.

## ***R&D Approaches***

### ***Experimentation***

The experimental focus of the proposal is the identification and measurement of the key parameters affecting the migration and permanent storage of CO<sub>2</sub>. A combination of flow-through (dynamic) and static (batch) experiments in which reservoir and caprock materials are exposed to scCO<sub>2</sub> will be used to determine relative permeability of CO<sub>2</sub>, brine, and oil; dominant modes of CO<sub>2</sub> migration via viscous fingering, wormhole formation, diffusion, etc.; mineral reactivity in reservoir rocks as it affects permeability, porosity, and permanent sequestration via precipitation of carbonates; and the permeability and integrity of the caprock used for hydrodynamic trapping and the cement grout used for sealing wells. These data will provide direct inputs for the proposed simulation studies and direct evidence for answering "how well will geologic sequestration work?".

*Flow-through (Dynamic) Experiments.* The injection of scCO<sub>2</sub> into core samples (Table 2) will be conducted at the Supercritical Fluids Facility (SCRUB) and C-INC to study 2-phase fluid interactions and at The Petroleum Recovery Research Center (PRRC) at New Mexico Tech for 3-phase fluid studies. High-pressure equipment and expertise resides at all locations for small-scale (10mL vessels) to large-scale (70L) experiments. In addition, the SCRUB is a unique facility with extensive analytical capabilities for exploring the interaction of scCO<sub>2</sub> with a wide-range of materials including cement grouts and rocks<sup>[8,9]</sup>. The PRRC is a national leader in studies of 3-phase (CO<sub>2</sub>-oil-brine) permeability relations in secondary oil recovery. In all cases, the cores will be characterized for initial permeability, porosity, and mineralogy. The cores will be sealed, and scCO<sub>2</sub> or scCO<sub>2</sub>-brine mixtures will be forced through the core. Permeability will

be determined from a measured pressure drop across the core length. Relative permeability will be determined from the relative rate of fluid movement in the cores. Measured changes in permeability will provide important constraints for model development. Viscous fingering will be evaluated from permeability measurements and from mineral reaction textures. In addition, we propose to permeate selected cores with organic CO<sub>2</sub>-soluble dyes and examine core-sections to determine CO<sub>2</sub> flow-paths. Fluid samples will be taken from taps installed along the length of the cores. The fluid chemistry will provide constraints for the development of the geochemical reaction models. In addition we propose to install *in-situ* high-pressure devices to monitor particle size, pH, conductivity, and solution chemistry.

*Long-term (Batch) Experiments.* The low temperature of most potential geologic reservoirs implies slow mineral dissolution and precipitation rates. To evaluate reactivity in scCO<sub>2</sub>-brine-reservoir-caprock system and identify key reactions for long-term performance calculations, a suite of batch experiments will be performed at the hydrothermal laboratory in C-INC. We maintain a set of flexible cell hydrothermal systems capable of experimental investigation of fluid-mineral reactions to greater than 350°C and 1kbar with *in-situ* fluid/gas sampling capability<sup>[4]</sup>. The interface between scCO<sub>2</sub> and brine in a carbon repository may exhibit large gradients in chemical potential and, consequently, will display the greatest reactivity. Experiments will probe these potential reaction fronts by evaluating reservoir and caprock core positioned at the boundary between brine and scCO<sub>2</sub>. Paired experiments will provide fluid samples to monitor reaction progress without perturbing the interface. Additional experiments will compare reactivity of reservoir and caprock with acidified brine (that coexists with scCO<sub>2</sub>) and directly in scCO<sub>2</sub>. Initial experiments may be of higher temperature (~200°C) and shorter duration (3-5 months) to leverage existing work<sup>[4]</sup> and to maximize reaction rates under realistic (but more extreme) conditions. Subsequent experiments at low temperature (~100°C or less) and long duration (up to 12-24 months) will track key reactions and rates identified in initial experiments and simulations.

*CO<sub>2</sub>-Induced Modification to Rock.* Changes in porosity and mineralogy of select cores will be evaluated by neutron scattering and tomography at LANCSE and UC Davis. These techniques will allow imaging of changes in porosity (e.g., wormhole formation) at scales greater than 25 µm and changes in mineralogy > 5%. Bulk changes in porosity and pore-size distribution will be evaluated from changes in mass of the core and with mercury porosimetry or an equivalent method. Qualitative and quantitative features of changes to porosity will be determined by using scanning electron microscopy (SEM) and image analysis. Post-experiment analysis of sections taken from the core and from solids will also be used to determine changes in mineralogy (via optical microscopy, SEM, and X-ray diffraction). These changes in mineralogy will be related to mineral reaction rates by normalization to surface area<sup>[10]</sup>. The permeability of CO<sub>2</sub>-trapping materials (shale and cement grout) both with and without fractures will be determined as a basis for calculating leak-rates from reservoirs. Cores of these materials will be examined for mineralogical changes, especially healing or opening of fractures. Compressive and tensile strengths will also be analyzed for these materials to determine the effect of mineralogical reactions on structural integrity. These data will augment our earlier work, showing that scCO<sub>2</sub> may be beneficial to cement grouts by decreasing permeability and increasing strength<sup>[9]</sup>. However, these experiments need to be expanded to include the effect of brines, the age of the grout, shale caprocks, and reservoir formations. In addition to the analysis of the material parameters, the collected effluents from all experiments will be characterized. Standard



properties of solution will be collected as outlined in the flow section with the ability to increase the range of data collection to elemental/chemical components.

*Field-Scale Analogues.* The Hobbs, NM and other field CO<sub>2</sub>-injection sites, and the Bravo Dome CO<sub>2</sub> reservoir, provide natural laboratories for validating the experimental and modeling results relative to encapsulation of CO<sub>2</sub> at geologic time scales. We will analyze the significant amount of data (e.g., formation pressure, fluid chemistry, injection/withdrawal history, core characterizations, etc.) that have been collected for these fields.

## ***Simulation***

The role of modeling and simulation will be to incorporate the experimental data to develop an understanding of the long-term fate of CO<sub>2</sub> in reservoirs. Simulations will also be used to identify important processes that need to be studied in detail with experiments. A two-pronged approach will be used to understand processes at multiple scales. Microscopic scale models will be used to simulate the physical and chemical interactions of CO<sub>2</sub>, reservoir fluids, and geological media and to derive constitutive relationships that control the flow behavior. These constitutive relationships will be used in the coupled macroscopic models for simulating long-term behavior of CO<sub>2</sub> in large-scale injection. Implementation of such models will require making full use of LANL's high-performance computing capabilities and resources.

*Modeling — Develop models for microscopic interactions.* The microscopic modeling involves simulating interactions of CO<sub>2</sub>, reservoir fluids, and geological media at the pore-scale. The crucial aspect of the microscopic modeling will be incorporation and validation of the thermodynamic and kinetic data derived in the experimental portions of this project and development of upscaling strategies for deriving effective physical and chemical parameters needed for macroscopic modeling. Innovative, hybrid models that combine continuum and microscale processes will be needed for describing sharp reaction fronts.

1) Develop pore network and LB models to gain insights into microscopic interactions. We will use an enhancement of the Lattice Boltzmann Permeameter (LBP) and pore network models for numerical modeling. The LBP technique was developed at Los Alamos and received an R&D 100 award in 1994. We will extend the model to account for diffusion and reaction in 3D pore geometries for multicomponent systems on the basis of LANL's prior experience in 2D LB simulation of surface reactions<sup>[11-15]</sup>. We will simulate fluid flow, transport, and reaction processes with these pore-scale models on real rock geometries obtained by neutron tomography and SEM image analysis from actual cores used for experiments and integrate with experimental results. On the basis of the microscopic simulations, we will be able to investigate the change of physical and chemical properties of the rock due to dissolution/precipitation. We will also study the effects of pore-scale structures and processes on macroscopic coefficients (e.g., permeability, dispersivity, and reaction coefficients) and identify key microscopic parameters (e.g., pore-size distribution, diffusivity, and surface reaction rates) and predominant processes that control the macroscopic quantities.

2) Develop upscaling strategies for deriving macroscopic kinetic and thermodynamic models for the interactions. Develop macroscopic constitutive models for multi-phase flows and experimentally measure the parameters characterizing these models. We will develop upscaling strategies for deriving macroscopic kinetic and thermodynamic parameters for the complex system. The upscaling techniques derived from a previous LDRD/ER project (#99025) for non-reactive flows in porous media<sup>[14,16]</sup> will be evaluated and modified to account for the interplay of convection, diffusion, reaction, and pore-geometry evolution for reactive flows.

3) Develop an innovative, hybrid approach for modeling the dynamic process of wormhole formation. Two distinct models for describing flow and transport in porous media will be combined to account for processes at the pore and continuum scales. This approach is a hybrid algorithm, similar in concept to existing schemes used in fluid mechanical applications near sharp discontinuities (shocks and boundaries). In regions occupied by reaction fronts, a pore-scale (LB) model will be employed to account for heterogeneities at the microscale through computer-generated pore geometries on the basis of measured statistical quantities. Away from dissolution fronts in regions outside the front regions, a continuum scale model will be used, whose parameters are either obtained from lab experiments or upscaled from pore-scale models. The two models will be coupled across an interface, separating the front and continuum regions by matching average pressure and flux. The inhouse computer code FLOTRAN<sup>[17]</sup> will be used for the continuum-scale formulation. FLOTRAN is a multi-phase, multicomponent reactive flow and transport model that is applicable to variably saturated, nonisothermal systems.

*Modeling – Develop macroscopic simulator for predicting fate of disposed CO<sub>2</sub>.* Macroscopic models will be developed to study behavior of CO<sub>2</sub> and host media when large amounts of CO<sub>2</sub> are stored in the host formations. The two main issues studied will be long-term fate of injected CO<sub>2</sub> and potential impact of escape of CO<sub>2</sub> due to failure of natural barriers or migration along natural/anthropogenic escape paths such as faults or wells.

1) Long-term fate of CO<sub>2</sub> and host environment. To understand long term fate of CO<sub>2</sub> and the host environment, it will be necessary to incorporate the coupled hydrodynamic, thermodynamic, and geochemical processes. For example in an oil reservoir, important interactions will include thermodynamic reactions among CO<sub>2</sub>, oil, and brine; geochemical interaction between CO<sub>2</sub> dissolved in brine and reservoir rock; and migration of CO<sub>2</sub> controlled by the physical characteristics of the reservoirs. We will develop models that will have the capabilities to represent all of these interactions. No numerical simulators are currently available that can effectively model simultaneous 3-phase fluid equilibria and geochemical interactions in oil reservoirs. We will utilize the existing codes in EES Division. These codes are developed to model flow and transport of contaminants in groundwater and have the capabilities required to represent geochemical interactions between CO<sub>2</sub>, brine and reservoir rocks. These capabilities will be updated to represent geochemical interactions within diverse mineralogies. The necessary information and data on effective surface area, kinetic rates, and reaction products will be collected from laboratory experiments as well as the literature. The codes will be updated to include thermodynamic models that represent interaction among CO<sub>2</sub>, brine, and oil (as a multi-component mixture) at reservoir pressure and temperature. Multi-phase flow models will be updated based on the experimental data on relative permeability functions. The constitutive models developed from the macroscopic simulations will be used to develop models for changes in porosity and permeability of reservoir rock. The numerical simulators will be validated against the experimental data, including, data from batch experiments (to validate the capabilities to simulate geochemical reactions); data from flow through experiments on core samples of reservoir rock (to validate the capabilities to simulate coupled flow and reaction behavior); and field-scale data, including the Hobbs experiment and Bravo Dome (to validate field-scale simulation capabilities). These simulators will then be used to perform long-term fate calculations. These calculations will be used to perform what-if scenarios, test different hypotheses, and identify areas where additional data/experimental work is needed.

2) Impact of escape of CO<sub>2</sub> due to failure in reservoir/well integrity or migration through natural/artificial pathways. The results of experiments with caprock and cement will be used to



enhance capabilities of the simulators to model the failure scenarios. Experimental data on geochemical interactions between shale and CO<sub>2</sub>/brine will be used to develop models for CO<sub>2</sub>/caprock interactions. Models for transport of CO<sub>2</sub> across the caprock including diffusion, flow/reactive transport through variably filled fractures/faults, and geochemical reactions with shales will be incorporated in the simulators. In addition, capabilities to model reaction of CO<sub>2</sub> with cement in plugged and abandoned wells in oil reservoirs will also be incorporated. These capabilities will be developed based on the experimental results as well as extensive prior work that has been performed on concrete. The models will be used to perform analysis of impact of escape of CO<sub>2</sub> due to breach in caprock or migration through natural pathways, including the rate and probability of escape to atmosphere. These calculations will be useful in identifying the type of monitoring capabilities required for verification of long-term sequestration.

### ***Potential Impacts and Institutional Benefits***

The proposed research addresses fundamental R&D issues associated with using geological carbon sequestration for mitigating global warming due to CO<sub>2</sub>. It directly supports LANL's mission "**to enhance global security by ... providing technical solutions to energy, environmental, infrastructure, and health security problems.**" Our research also supports LANL institutional Goal #5, "to establish a major LANL initiative in civilian science and technology and a new program from DOE." Geologic sequestration is being recognized (e.g., by NCCTI) as the most immediate, viable option to mitigate global warming. Consequently, programs in this field have tremendous potential for growth in the near future. The current DOE carbon program emphasizes field demonstrations and less fundamental science, and the whole field of carbon sequestration science/engineering is still in its infancy. Our proposal will provide the fundamental data and modeling tools necessary to predict the long-term fate of CO<sub>2</sub> with geologic sequestration. This LDRD will help to develop LANL's position as the lead research facility in geologic sequestration. Due to the importance of CO<sub>2</sub> sequestration research, we anticipate publishing the results of these studies in the highest impact journals such as *Science* and *Nature*, as well as leading journals in the fields of geochemistry, hydrology, and the physics of fluids. The components of this project (e.g., coupled multi-phase flow and reaction processes, drilling and injection, etc.) are also part of the essential skills required to maintain LANL's underground containment science, petroleum engineering, and environmental science/engineering capabilities.